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# A PRELIMINARY INVESTIGATION FOR THE USE OF DIGITAL GAMMA-GAMMA COINCIDENCE SPECTROMETRY TO DETERMINE $^{239}$PU

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Abstract. ***Gamma-ray spectrometry is one of the most effective ways to determine the activity of $^{239}$Pu, depending on activity levels. However, often high backgrounds in complex spectra with low amounts of $^{239}$Pu can increase detection limits. The effectiveness of the use of gamma-gamma spectrometry in the characterization of $^{239}$Pu was studied for the first time. Using the XIA Pixie-16 digital pulse processor, the gamma-gamma system was studied to establish unshielded background radiation, source to detector distances, Poisson statistics and gating of several gamma-rays from $^{239}$Pu. The results clearly show that the distance between gamma ray spectrometers has a much bigger effort in counting statistics for the gamma-gamma coincidence detectors than a single detector. In addition, measurements demonstrated the background radiation to be virtually negligible, meaning gamma-gamma spectroscopy can still be very effective without requiring the usual lead shielding. Other measurements were taken to assure Poisson statistics were attained in the digital system. Preliminary measurements revealed 3 orders of magnitude background reduction for the measurement of $^{239}$Pu using gated gamma-rays.***

**Key words**: *Background reduction, gamma-gamma coincidence spectrometry, PIXIE XIA, Poisson statistics*

## 1. Introduction

Gamma ray spectrometry with a single germanium detector has been previously used for the characterization of the low-energy gamma rays of $^{239}$Pu [1]. In addition, it has been used for determination of $^{239}$Pu/$^{241}$Pu ratios in uranium-plutonium mixed oxides [2]. However, the background when using a single unshielded gamma-ray detector can be on the order of thousands of counts, depending on the counting times, energy in the spectrum, and detector efficiency causing increased detection limits. One method to reduce the background, and therefore the detection limits, is the use of both active and passive shielding [3]. Another option is the use of gamma-gamma coincidence, which has been used for $^{134}$Cs [4], among other isotopes, but not for $^{239}$Pu. Gamma-gamma coincidence has been proven capable of significantly reducing background radiation, which is advantageous in the characterization of an isotope with a complicated decay scheme, like $^{239}$Pu.

The use of gamma-gamma coincidence is valuable when used on radionuclides that decay to an excited state, then immediately decay to a lower energy state. In this situation, the two gamma rays emitted from the decays would be in coincidence, and would show up in gamma-gamma coincidence detection. For this reason, the use of coincidence has been demonstrated to significantly reduce background counts [5]. Lawrence E. Wangen et al. found that the use of gamma-gamma coincidence produces a count reduction of 3 to 4 magnitudes when measuring selenium ($^{75}$Se) in environmental standards using neutron activation analysis [6]. Gamma-gamma coincidence counting allows for lower detection limits, particularly in conjunction with the use of energy gating. Truong Van Minh et al. performed a comparison of normal and coincidence gated spectra and determined an improvement in both peak to background ratio and detection limits with the use of selenium [7]. J. Konki et al. have also produced evidence demonstrating lower detection limits and reduced background counts with the use of gamma-gamma coincidence [8]. In addition, the use of coincidence techniques has been demonstrated to be effective in deconvolution of overlapping peaks, particularly in $^{76}$As, $^{160}$Tb, and $^{169}$Yb [9].

## 2. Experimental

The gamma-gamma coincidence system utilized two high-purity germanium detectors. One of the HPGe detectors is a Canberra CG2518 which has a relative efficiency of 25% and an energy resolution of 1.95 full width half maximum (FWHM) at 1332 keV of $^{60}$Co. The second HPGe detector is also a Canberra CG2519 which has a relative efficiency of 15% and an energy resolution of 1.9 FWHM at 1332 keV. The detectors are linked to a digital XIA PIXIE-16 module with digital spectrometry and coincident data acquisition capabilities [10].

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To determine the comparison of unshielded background counts detected with coincidence versus singles, a spectrum was taken using both systems with no source for 5 hours as seen in Figure 1.

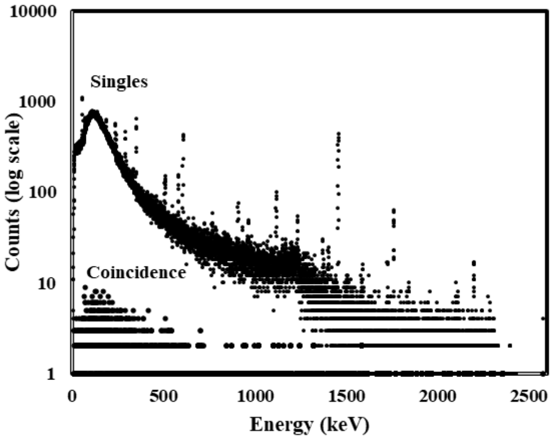


Figure 1. HPGe Coincidence and Singles Background Spectra.

This comparison proves that the use of gamma-gamma coincidence significantly reduces the background counts without the use of any shielding.

A $^{60}Co$ source was measured in both singles and coincidence modes at various distances from the face of the detectors, and the 1332 keV net area counts were compared. Using a single detector, the net counts decreased by an order of magnitude at a source to detector distance of 10 cm. Using the two detectors in coincidence, however, results in a decrease of two orders of magnitude in the counts at a source to detector distance of 10 cm. The singles and coincidence data at varying distances are displayed below in Figure 2 and Figure 3.

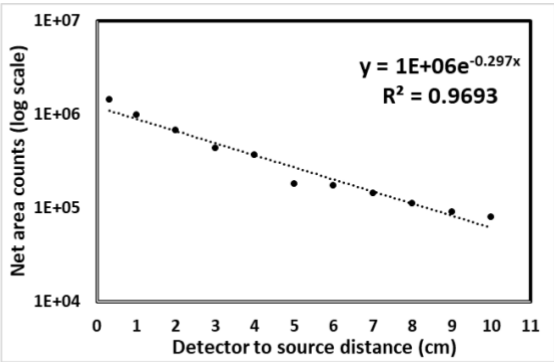


Figure 2. HPGe Singles 1332 keV Net Area Counts at Several Source to Detector Distances.

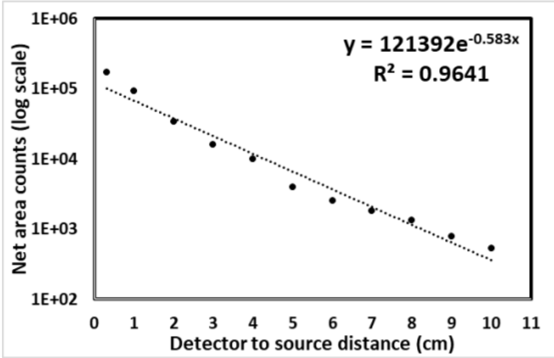


Figure 3. HPGe Coincidence 1332 keV Net Area Counts at Several Source to Detector Distances.

From the figures above, it can be seen that the reduction in counts with the use of coincidence becomes more significant at greater distances. This is to be expected, as the number of gamma rays that reach both detectors within 80 nanoseconds, the coincidence time window, is inversely related to source to detector distance.

### *2.1. Characterization of PIXIE-16 System*

After determining the value of the use of gamma-gamma coincidence for background net count reduction, a characterization of the PIXIE-16 system was performed. A verification of Poisson properties of the system was done by placing a $^{60}Co$ source equidistant between the two detectors and performing 2500 runs on the system. Each run was 200 seconds, and a custom Python code was used to ensure the elimination of human error. Both the singles and coincidence data were analyzed from the PIXIE-16 system, and produced the distributions in Figure 4 and Figure 5 below.

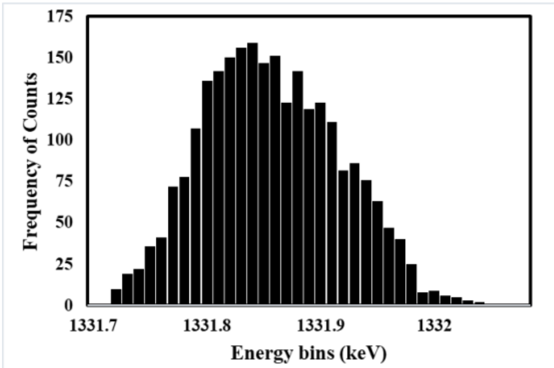


Figure 4. Poisson Distribution Single HPGe 2500 Runs with 200 s Counting Time for 1332 keV $^{60}Co$ Gamma.

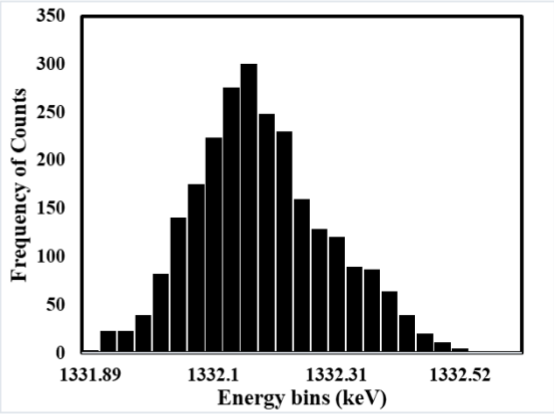


Figure 5. Poisson Distribution Coincidence HPGe 2500 Runs with 200 s Counting Time for 1332 keV $^{60}$Co Gamma.

From the datasets generated from singles and coincidence data in Figures 4 and 5, the PIXIE-16 system abides by Poisson distributions.

### *2.2. Advantages of Coincidence Counting for $^{239}$Pu*

The value in using $^{239}$Pu in the gamma-gamma coincidence system can be determined from its decay scheme. A simplified version of the decay scheme of $^{239}$Pu is shown in Figure 6, with only the usual major gamma rays displayed.

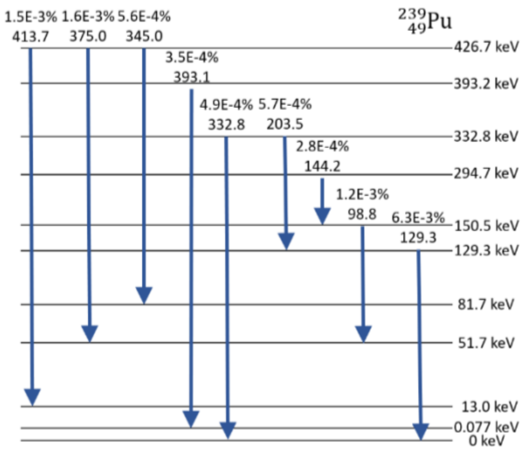


Figure 6. Major Gammas Belong to the $^{239}$Pu Decay Scheme.

In the decay scheme, there are multiple gamma rays that are in coincidence with each other, including the 144 keV and 98.8 keV gamma rays, and the 203 keV and 129.3 keV gamma rays. Therefore, the use of gamma-gamma coincidence on $^{239}$Pu would be effective due coincident gamma rays.

### *2.3. Coincidence Gating of $^{239}$Pu*

Once it was determined that $^{239}$Pu has gamma rays in coincidence, experimental data was evaluated to observe the coincidences. Figure 7 includes an overlay of singles and coincidence spectra of a $^{239}$Pu foil electrodeposited on nickel with an activity of 1.81E6 Bq.

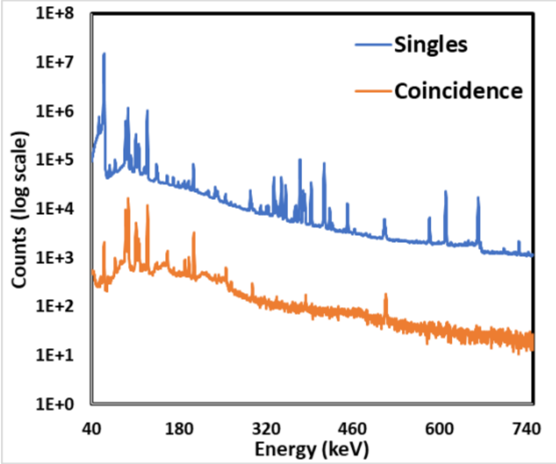


Figure 7. $^{239}$Pu Foil Singles and Coincidence with 97-hour Live Time Count

The comparison of the singles and coincidence data was then used to evaluate the gamma rays in coincidence. From Figure 7, it can be seen that the most gammas in coincidence are in the range of 94 keV to 129 keV, with almost no gammas in coincidence above that range. Therefore, several gammas within the range were analyzed further by energy gating.

The gating was done by the use of energy gating software, in which the energy of interest at detector 1 was specified by the user after performing a run with the two detectors touching the $^{239}$Pu source. The software then found all energy peaks in detector 2 that were in coincidence with the specified energy. This process was performed on the 94 keV, 103 keV, 111 keV, and the 129 keV energy peaks. The energy gated spectrum for 129 keV is shown below in Figure 8. The coincidence spectrum is overlaid on the energy gated spectra for reference.

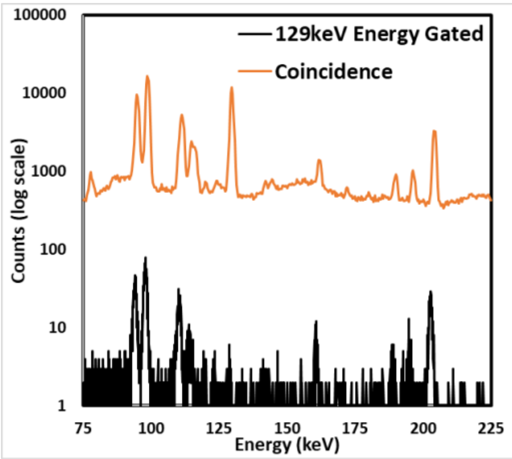


Figure 8. 129 keV Energy Gated and Coincidence Spectra Overlay of $^{239}$Pu.

The gated spectrum demonstrates experimentally the gamma rays 94, 98, 111, 114, and 203 keV are in coincidence with the 129 keV gamma. In addition, the gated spectrum has a background count reduction of 3 orders of magnitude relative to the coincidence data, and 5 orders of magnitude reduction relative to the singles data.

To quantify the background reduction, a comparison of the net counts of the 203 keV peak in the singles, coincidence, and gated spectra was done. The 203 keV peak is indicated by the red box in Figure 9 below, and the net counts are shown in table 1.

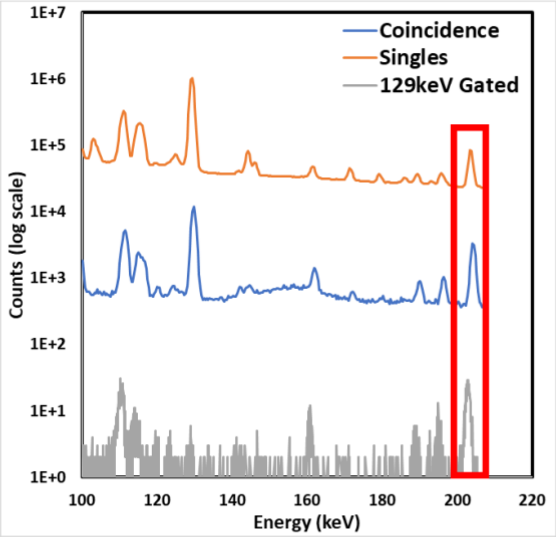


Figure 9. Singles, Coincidence, and 129 keV Gated Spectra Comparison of 203 keV Peak

Table 1. Net counts in 203 keV peak in singles, coincidence, and gated spectra

| 203 keV Peak | Net Area | Ratio of Net Area to Gated Net Area |
|---|---|---|
| Singles | 258419±790 | 364 |
| Coincidence | 12727±137 | 18 |
| 129 keV Gated | 710±29 | 1 |

The ratio of net counts of the singles to the energy gated data at 203 keV is 364, from Table 1. This reduction of greater than two orders of magnitude of the net counts indicates that gamma-gamma coincidence can be used with the absence of shielding.

## 3. Conclusion

The use of gamma-gamma coincidence, without any shielding, reduces background counts by two orders of magnitude. The XIA PIXIE-16 system maintains Poisson statistics for coincidence and singles counting, and therefore, data from the system can be considered accurate. Gamma-gamma coincidence can be used for $^{239}$Pu, as the isotope has several gamma rays in coincidence, including the 94, 98, 115, and 129keV gammas. The use of energy gating, in addition to coincidence, further reduces the background by 3 more orders of magnitude and is an efficient method to experimentally determine the gamma rays in coincidence with a particular gamma ray.

The lack of shielding required for background reduction, in addition to the numerous gamma rays of the isotope in coincidence make this method ideal for verification purposes of $^{239}$Pu. At present, gamma-ray spectroscopy is used for determination of plutonium isotopic measurements [11]. However, gamma-gamma coincidence can be used to determine the plutonium isotopic composition more effectively, even in samples with high americium content.

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